Terahertz dynamics of quantum materials

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What are quantum materials?

- Materials where ***emergent*** properties appear (cannot be predicted by properties of single atoms)

- ***Examples:***
  - Magnetism
  - Superconductivity
  - Strongly correlated phenomena
  - Topologically protected states

- Interplay of many degrees of freedom important (lattice, spin, electrons): many occurs at THz frequencies (meV energies)

Why ultrafast x-ray probes are important?

- femtoseconds are the time-scales of electronic and atomic motion

- paradigm shift where we can look ***directly*** at quantum mechanics in time and space

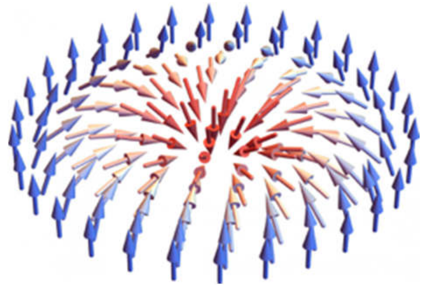
- new fundamental understanding of open problems in physics of quantum materials

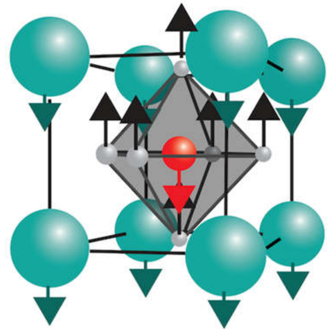
Why high rep-rate XFELs for condensed matter?

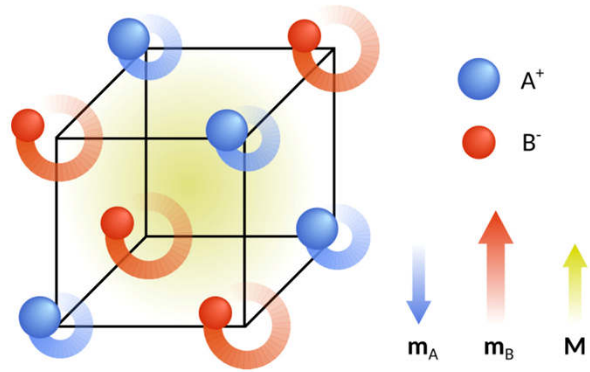
- Ultrafast imaging with nanometer resolution
- Electron and spin spectroscopy with femtosecond resolution
- High-sensitivity time-resolved x-ray diffraction

Outline

- ***Three scientific cases:***
  - Imaging of ultrafast magnetism
  - Nonlinear phonon dynamics
  - Dynamical multiferroicity
- A "dream" SASE4/SASE5 implementation at European XFEL







Why magnetism and why ultrafast?

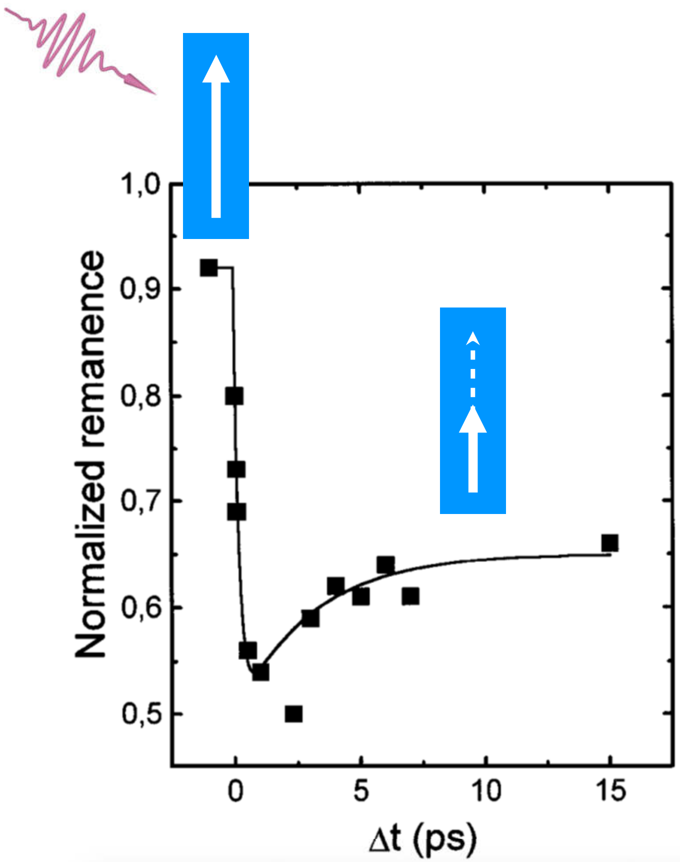
Why the Future of Data Storage is (Still) Magnetic Tape

Disk drives are reaching their limits, but magnetic tape just gets better and better

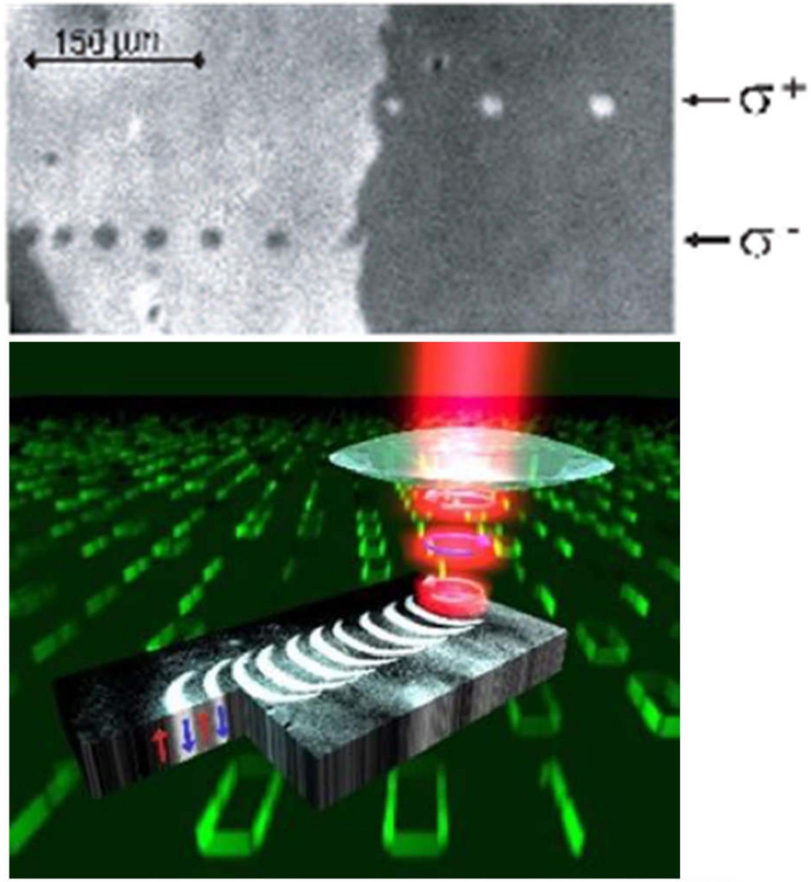
By **Mark Lantz**

- 90%+ storage in data-centers worldwide is magnetic (cheap and reliable)
- Bottle-neck for computing speed is storage
- Data-centers are very energy-hungry (most energy goes into heat and cooling):
  - 5% world electricity, 2-3% emission (now)
  - 20% world electricity (2025)

Ultrafast magnetism

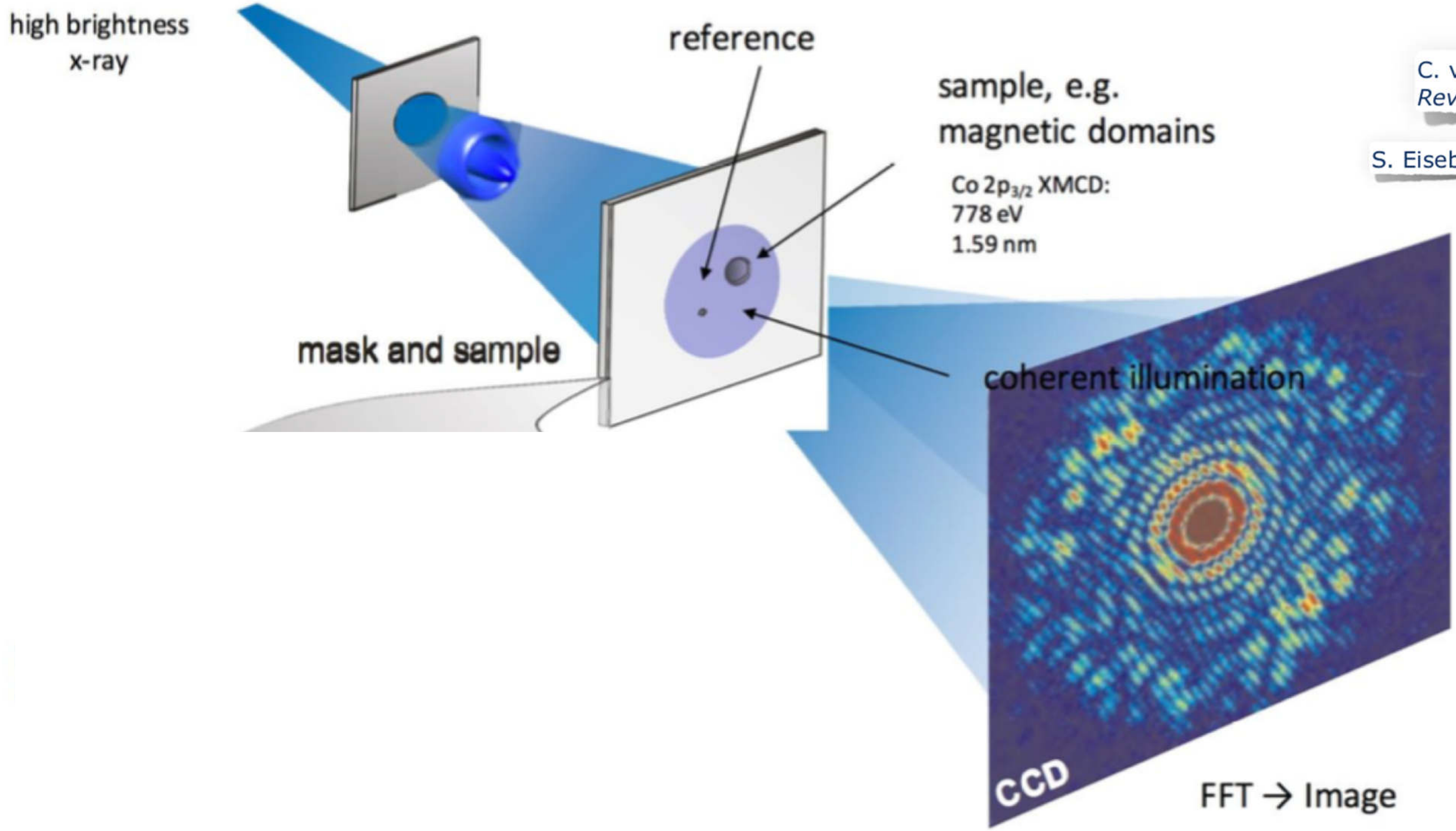


Beaurepaire, E, J-C Merle, A Daunois, and J-Y Bigot. "Ultrafast Spin Dynamics in Ferromagnetic Nickel." *Physical review letters* **76**, 4250 (1996)



Stanciu, C D, F Hansteen, A V Kimel, A Kirilyuk, A Tsukamoto, A Itoh, and Th Rasing. *Physical Review Letters* **99**, 047601 (2007)

X-ray magnetic holography: some details

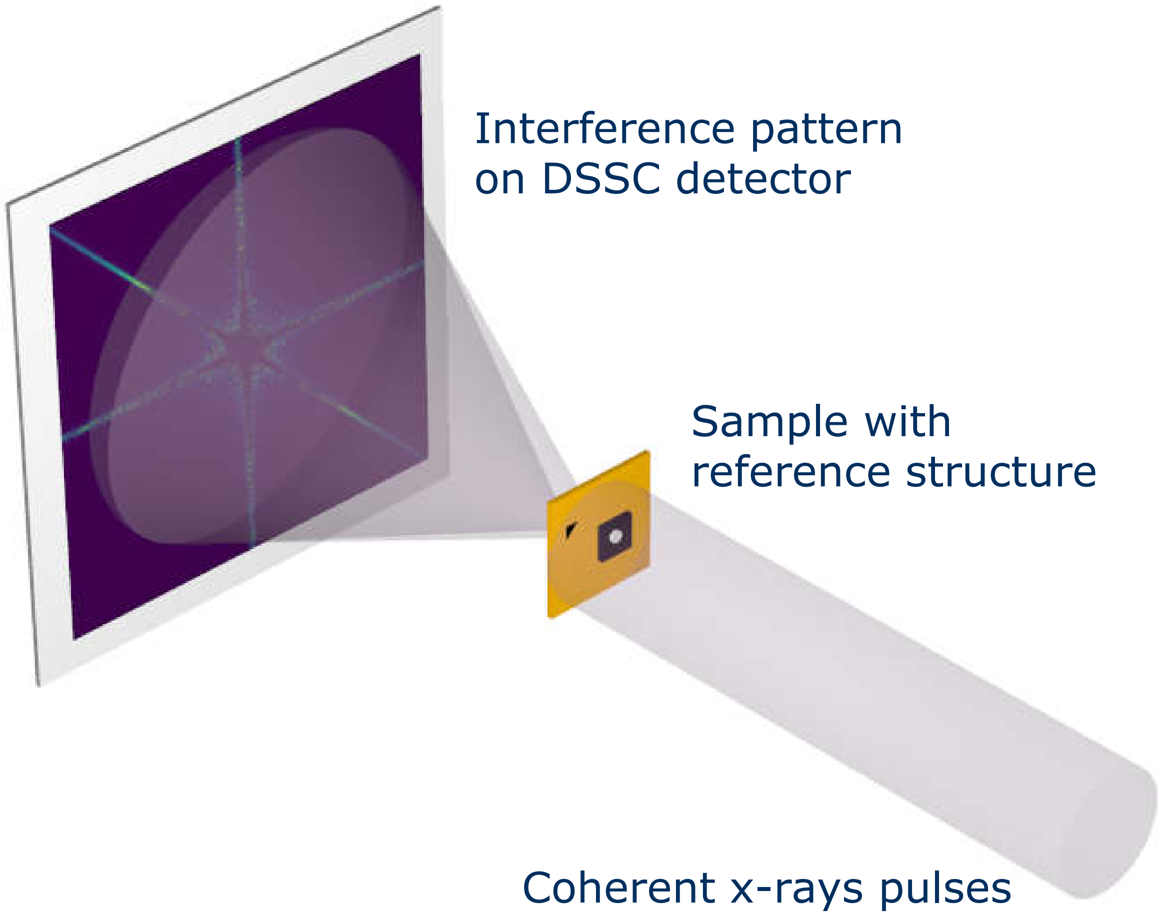


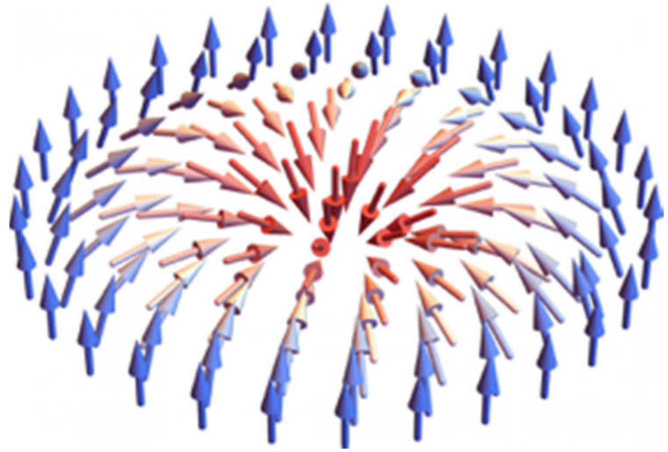
C. von Korff Schmising et al, *Phys. Rev. Lett.* **112**, 217203 (2014)

S. Eisebitt *et al.*, Nature **432**, 885 (2004)

Community proposal at European XFEL - accepted

X-ray holography of ultrafast magnetism: femtosecond movies at the nanoscale

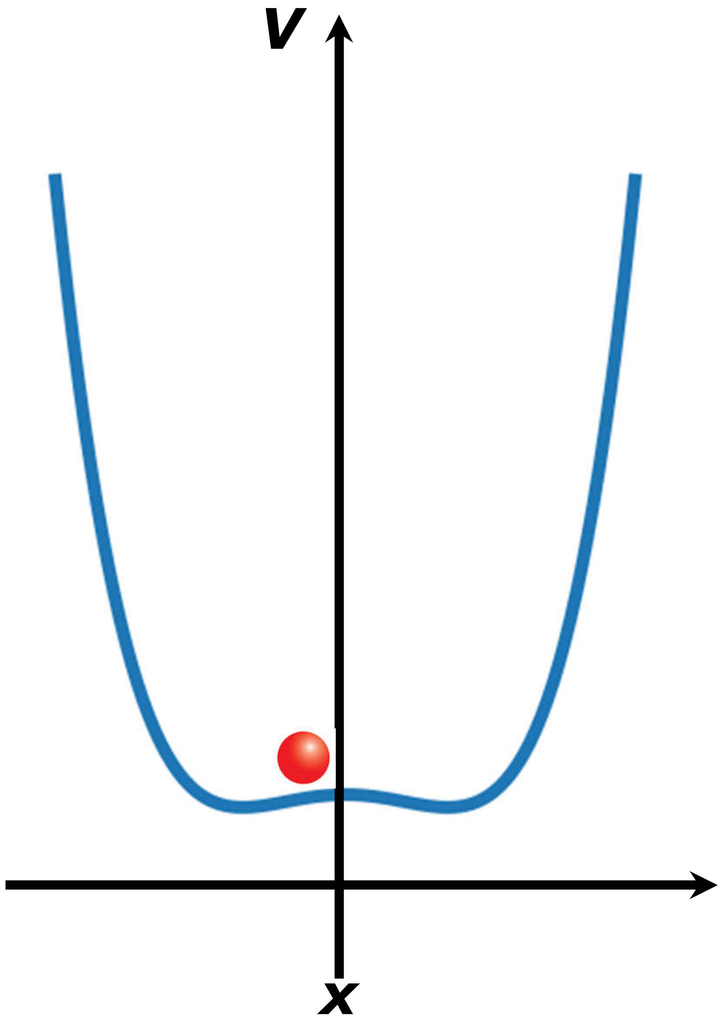


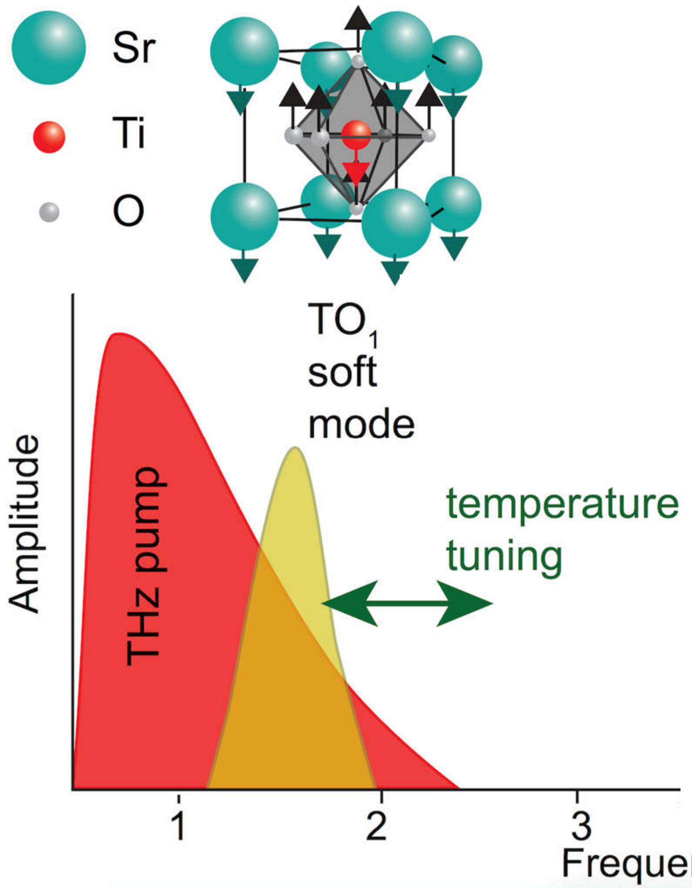


Skyrmion

S. Bonetti et al, XFEL Proposal 2222,
beam time allocated May 2019

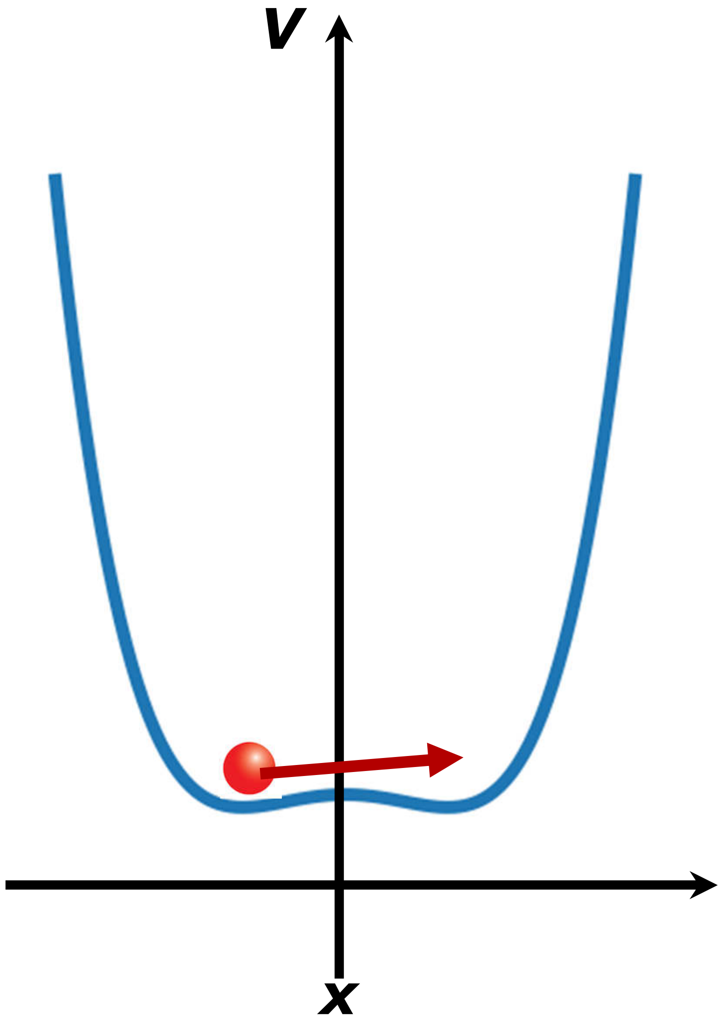
$SrTiO_3$ (STO): perovskite with soft phonon mode



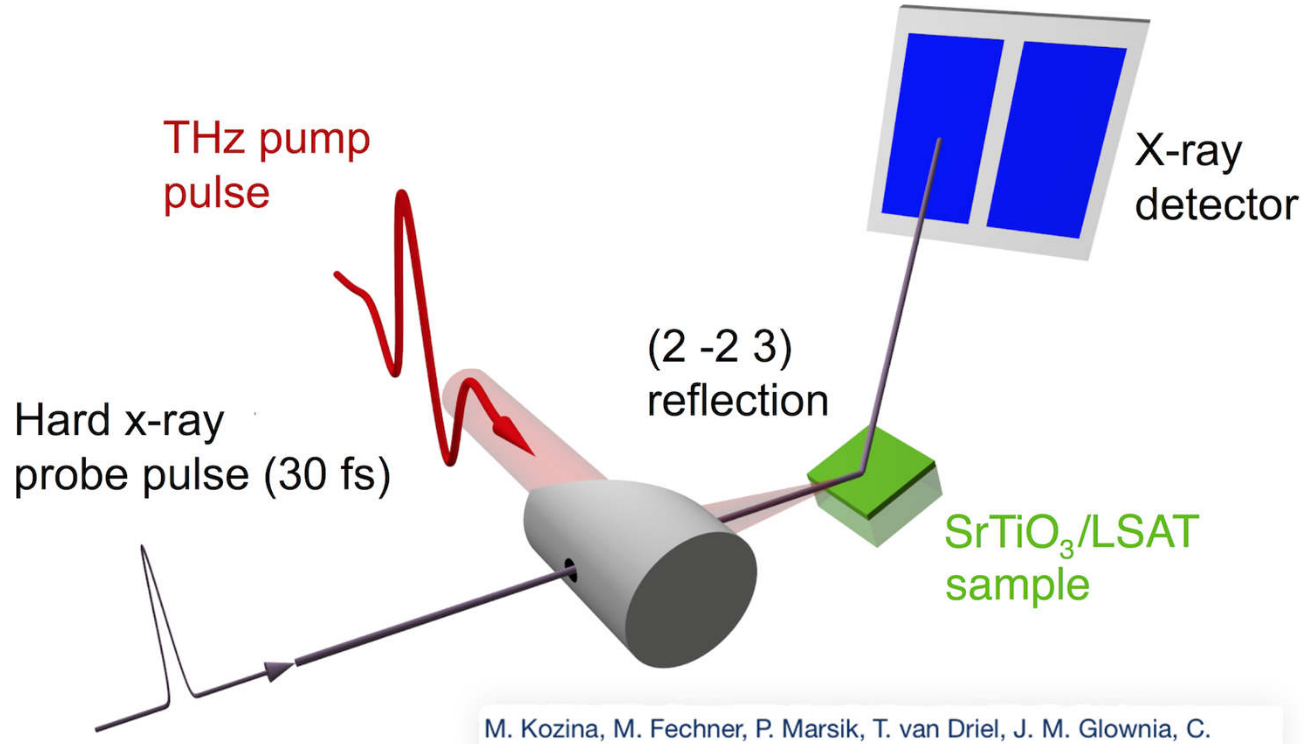


M. Kozina, M. Fechner, P. Marsik, T. van Driel, J. M. Glownia, C. Bernhard, M. Radovic, D. Zhu, **S. Bonetti**, U. Staub, M. Hoffmann, *Nature Physics in press (2019),* arXiv:1807.10788 (2018)

Idea: drive the soft phonon and probe the atomic motion *directly*



Collaboration with Matthias Hoffmann and Mike Kozina, Stanford / SLAC



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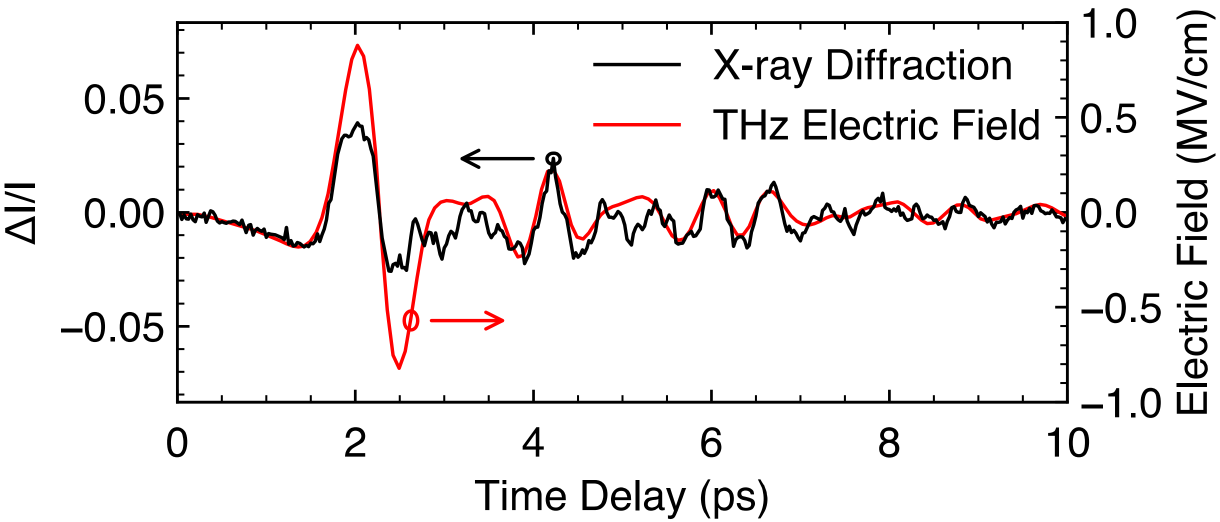
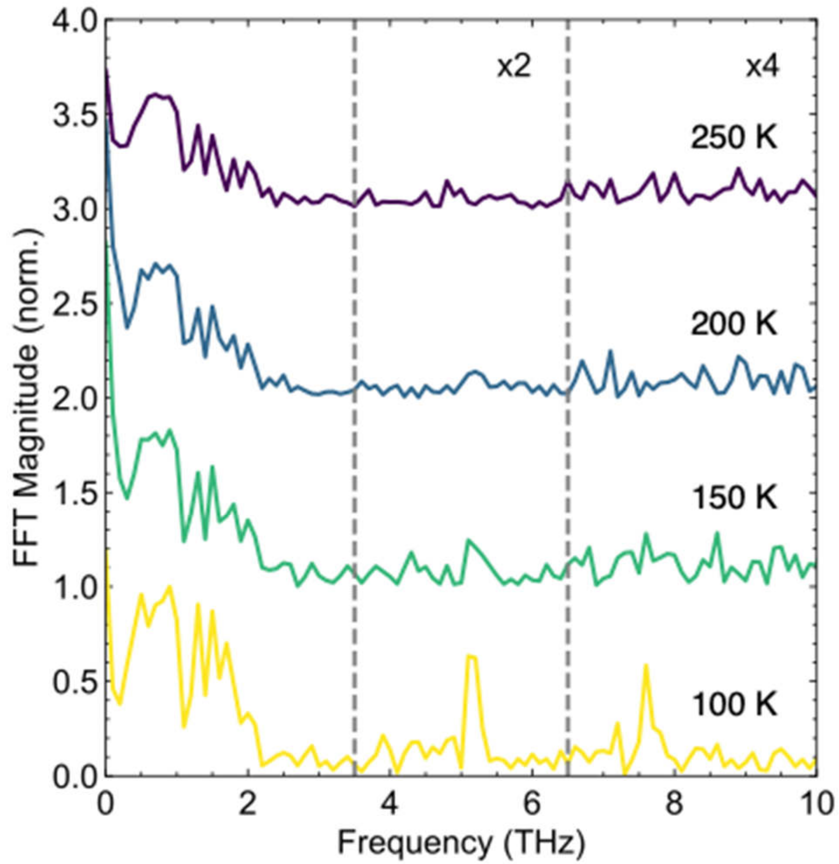
Phonon up-conversion





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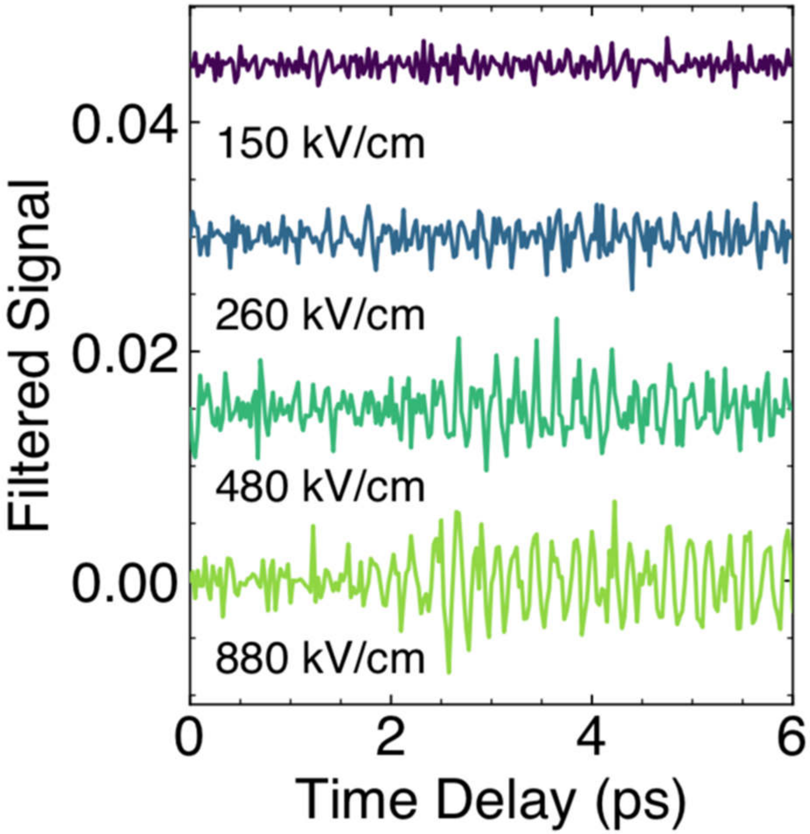
Mechanism: nonlinear (anharmonic) coupling

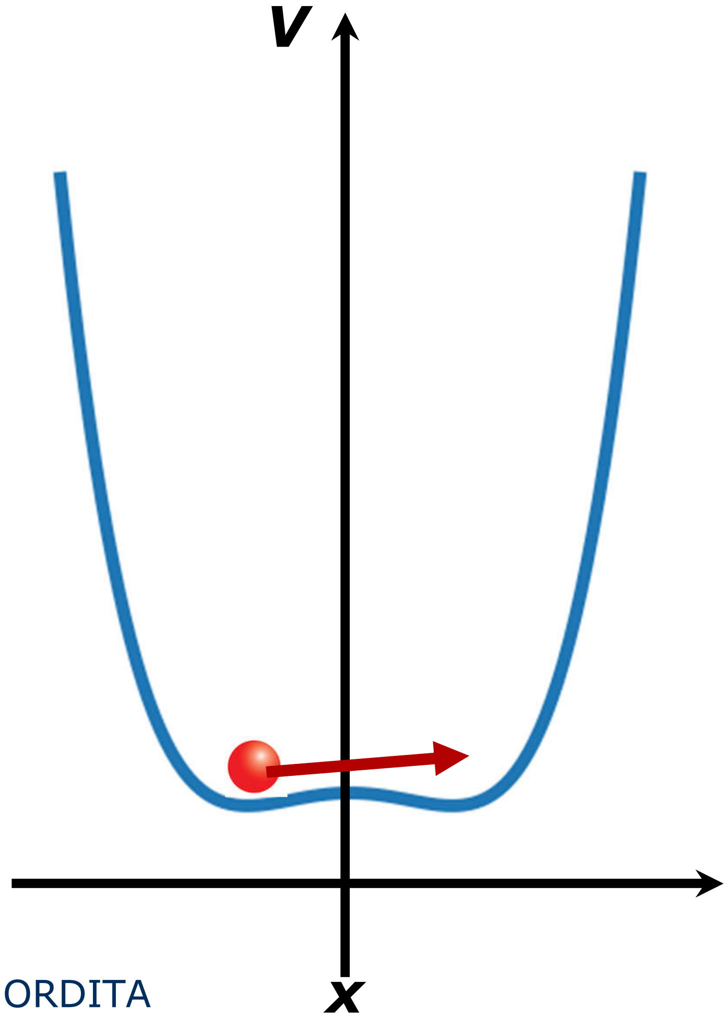


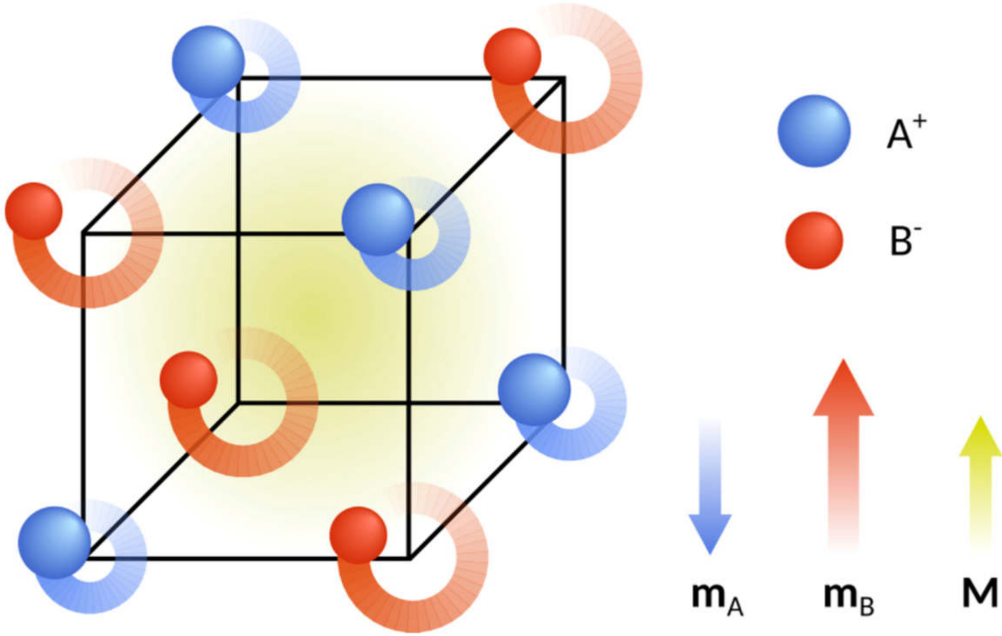
- 1.3 pm motion (15% of full displacement in the ferroelectric $BaTiO_3$ at RT)
- Drive 1 THz mode (TO_1), excite 5 THz mode (TO_2)
- Excite also "silent" mode at 8 THz (TO_3): access of symmetry-forbidden modes via nonlinear coupling

M. Kozina, M. Fechner, P. Marsik, T. van Driel, J. M. Glownia, C. Bernhard, M. Radovic, D. Zhu, **S. Bonetti**, U. Staub, M. Hoffmann, *Nature Physics in press (2019),* arXiv:1807.10788 (2018)

Next step: quantum paraelectric phase (<35 K) and dynamical multiferroicity

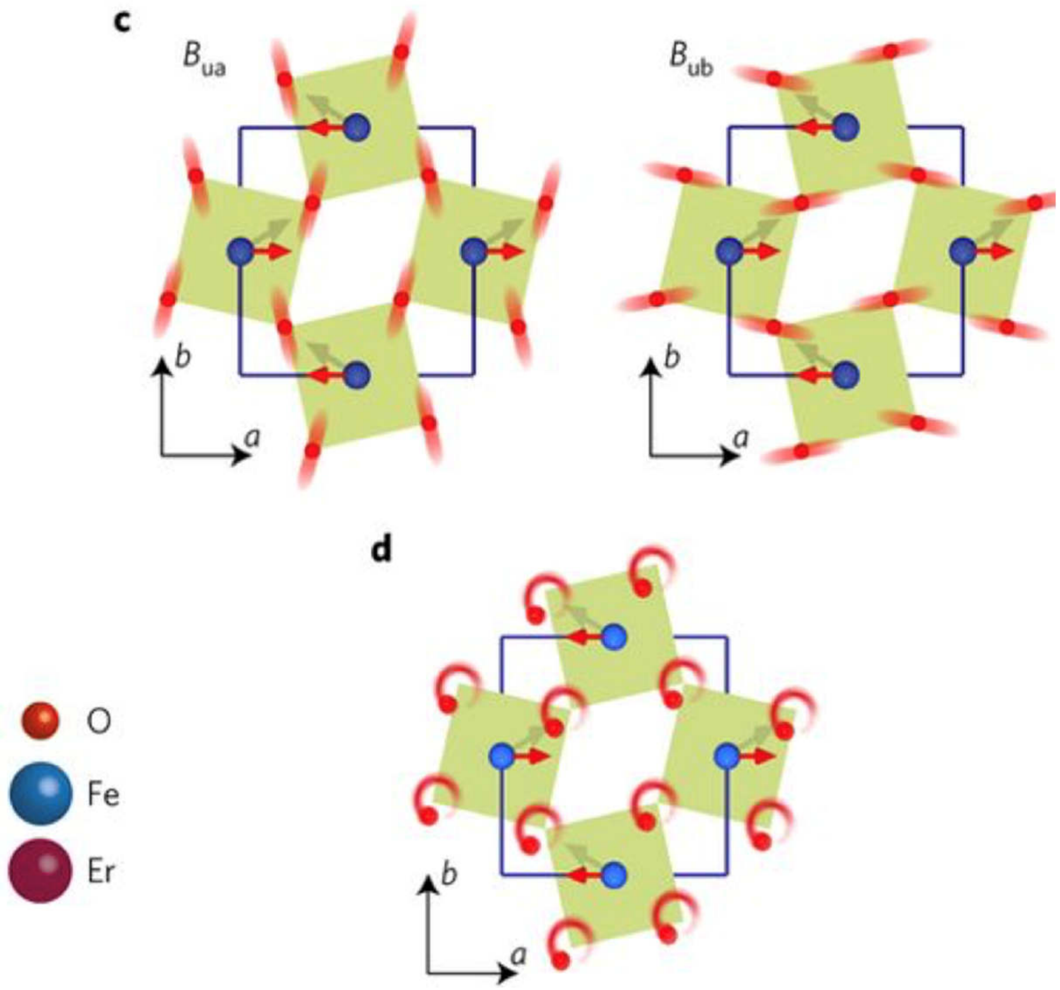
Collaboration with
Sasha Balatsky, NORDITA





D. M. Juraschek, M. Fechner, A. V. Balatsky, and N. A. Spaldin, *Phys. Rev. Materials* **1**, 014401 (2017)

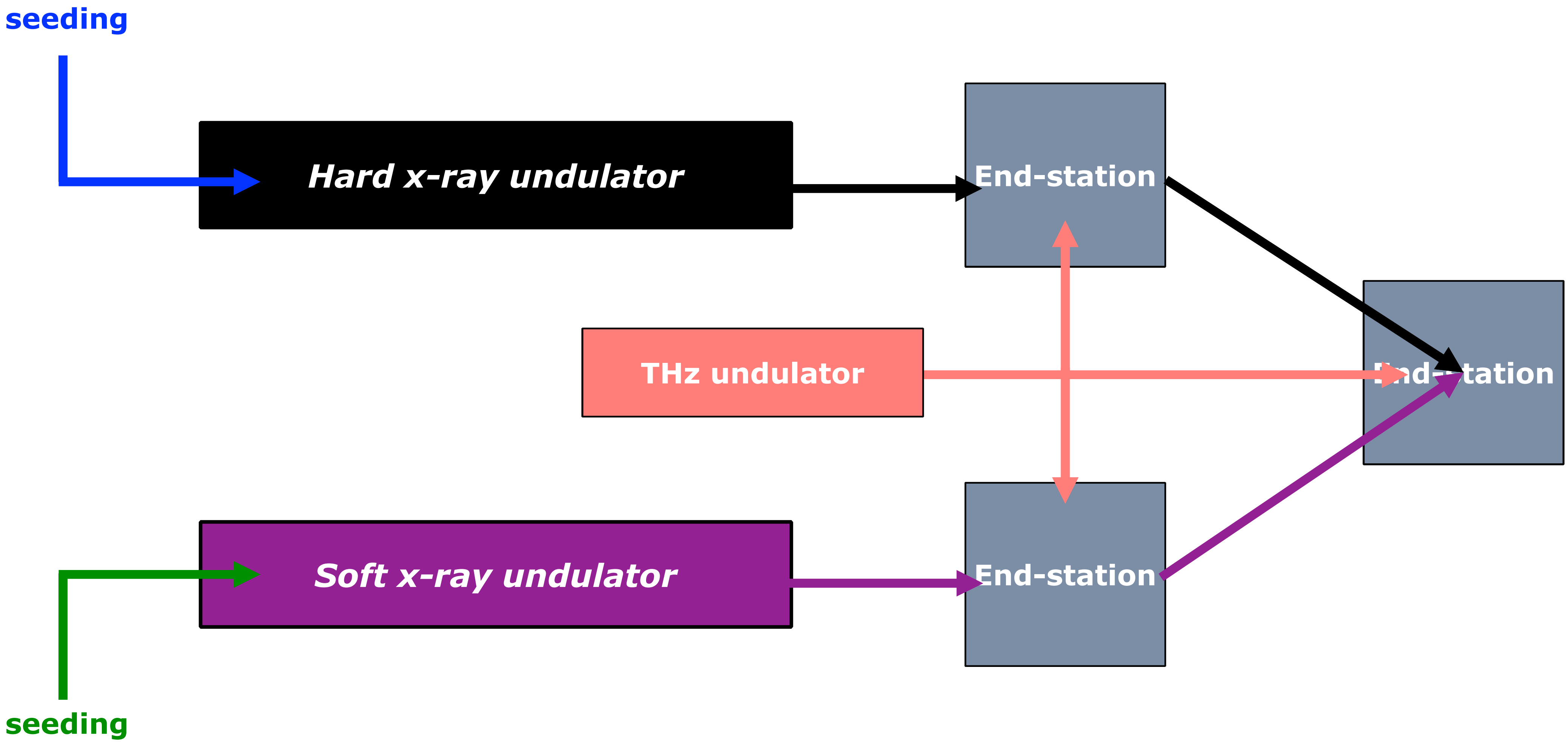
Effective magnetic field from driven phonons



- Two orthogonal phonons at f_1 = 16 and f_2 = 17 THz
- Effective circular orbit of the ions, at the beating frequency f_1 - f_2, resonant with magnon in the system
- Effective magnetic field produced

T. Nova et al, *Nature Physics* **13**, 132–136 (2017)

Dream set-up for quantum materials



High-rep rate machine goals

- Simultaneous spectroscopy and diffraction (correlated materials, e.g. dynamical multiferroics, electron-phonon coupling in superconductors)

- Combined hard and soft x-ray ultrafast imaging (antiferromagnets with THz resonances)

- Dirac materials?

Thank you for your attention!